Formation of Fe(III) (hydr)oxides from Fe(II) sulfides: Implications for akaganeite detection on Mars

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Akaganeite (β-FeO(OH,Cl)) is a Fe(III) (hydr)oxide with a tunnel structure typically occupied by chloride. This mineral was first proposed to be present in Meridiani Planum and Gusev crater on Mars and akaganeite was subsequently detected by X-ray diffraction in Yellowknife Bay and Vera Rubin Ridge locations in Gale crater. Akaganeite in Gale crater has been proposed to form from Fe(II) sulfides but formation conditions remain unconstrained.

We investigated akageneite formation by oxidative alteration of natural Fe(II) sulfide pyrrhotite exposed to HCl and oxidation-hydrolysis of Fe(II) HCl-leached from pyrrhotite at initial pH_0 1.5, 2, 3, and 4. X-ray diffraction and Mössbauer analyses revealed formation of poorly-crystallized akageneite in oxidative alteration experiments. Air-exposure of the HCl-reacted dry pyrrhotite led to an increase in akageneite formation and precipitation of Fe(II) hydrated sulfates, goethite, and hydronium jarosite. Iron(II) oxidation-hydrolysis was sensitive to Si dissolved from phyllosilicates in one pyrrhotite sample. Akaganeite and goethite formed at pH_0 1.5 and 2 with akageneite more abundant at dissolved Si/Fe ratio of 0.08 and goethite more abundant at Si/Fe of 0.01. Akaganeite formed together with hematite, ferrihydrite, and goethite at pH_0 3, and formation was suppressed at pH_0 4. Well-crystallized akageneite precipitated at pH_0 1.5 while akaganeite of poorer crystallinity formed at pH_0 2 and 3.

Akageneite in Gale crater could form from sulfides by both mechanisms: oxidative alteration of sulfide minerals and oxidation-hydrolysis of leached Fe(II). The processes likely occurred during late diagenetic events triggered by interactions of acidic Cl-bearing groundwater with Fe(II) sulfides. Akaganeite in Yellowknife Bay, Gale crater could have formed by Fe(II) oxidation-hydrolysis either as a sole Fe(III) (hydr)oxide at pH<2 or along with ferrihydrite and hematite at 2<pH<4 in Si-enriched conditions. Akaganeite formation at Vera Rubin Ridge, Gale crater could have occurred through oxidative alteration of sulfides in Cl-bearing pH 1.2-1.5 solutions. The presence of well-crystalline akageneite in Vera Rubin Ridge indicates that Fe(II) oxidation-hydrolysis contributed to akageneite formation.

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